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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US83/01713 <b>(22) International Filing Date:</b> 31 October 1983 (31.10.83) <b>(31) Priority Application Number:</b> 446,939 <b>(32) Priority Date:</b> 6 December 1982 (06.12.82) <b>(33) Priority Country:</b> US  <b>(71) Applicant:</b> HUGHES AIRCRAFT COMPANY [US/US]; 200 North Sepulveda Boulevard, El Segundo, CA 90245 (US). <b>(72) Inventor:</b> MUELLER, William, A. ; 1476 Sunshine Drive, Glendale, CA 91208 (GB). <b>(74) Agents:</b> LACHMAN, Mary, E. et al.; Hughes Aircraft Company, Post Office Box 1042, Bldg. C2, M.S. A126, El Segundo, CA 90245 (US).		<b>(81) Designated States:</b> AT, BE (European patent), CH (European patent), DE (European patent), FR (European patent), GB (European patent), JP, SE (European patent).  <b>Published</b> <i>With international search report.</i> <i>With amended claims and statement.</i>
<b>(54) Title:</b> METHOD OF CLEANING ARTICLES USING SUPER-CRITICAL GASES  <b>(57) Abstract</b>  In the fabrication of a variety of articles, organic contaminants become affixed to the surfaces of these articles and must subsequently be removed. The disclosed method is effective for the removal of organic contaminants from a variety of articles, and is efficient to enable the rapid removal of such contaminants without damage to the article. The method has special utility in the cleaning of components used in aerospace applications. In the disclosed method, a structural component bearing the contaminant is contacted in a pressure vessel with a gas under super-critical conditions of temperature and pressure whereby the contaminant is absorbed by the gas. The gas, having the contaminant absorbed therein, is then purged from the vessel to obtain the cleaned component.		

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METHOD OF CLEANING ARTICLES  
USING SUPER-CRITICAL GASES

BACKGROUND OF THE INVENTION

1     1. Field of the Invention

          This invention relates in general to a method of removing contaminants from articles and, in particular, to a simple, rapid and effective method of removing  
5     from the surface and interstices of a solid article a variety of contaminants with which the article may possibly have come in contact during its manufacture. More specifically, the present invention relates to a method of removing organic contaminants from such  
10    articles using gases in the super-critical state.

2. Description of the Prior Art

          Components and materials used in the manufacture of instruments for aerospace applications must be free  
15    from contaminants. The presence of trace amounts of contaminants in components of precision instruments used in space vehicles which ordinarily do not interfere with the operation of these devices on earth, manifest themselves under the conditions of outer space and  
20    interfere with the accurate, normal operation of these sensitive devices. Thus, it is critical that the components be free of any and all contaminants, particularly organic contaminants. The contamination



1 of the component may consist of saponifiable materials  
such as oils as well as non-saponifiable materials such  
as resins. Components formed from metal or synthetic  
plastic materials may contain gaseous or vaporizable  
5 contaminant residues from the manufacture and processing  
of the metal such as uncured prepolymers, release  
agents and unreacted monomers used in the processing  
of these materials.

To effect the required level of cleaning of the  
10 materials used in the manufacture of components which  
meet government standards for cleanliness, the art has  
developed cleaning processes for these materials utilizing  
high vacuum, e.g.,  $10^{-5}$  torr (millimeters of mercury or  
mmHg) and elevated temperatures up to  $250^{\circ}\text{C}$  to remove  
15 absorbed and adsorbed organic contaminants from the  
materials. This cleaning technique, referred to in the  
art as "thermal vacuum cleaning", is not completely  
satisfactory in that the cleaning process must be  
carried out in an expensive and complex high vacuum  
20 system which normally requires about fifteen hours to  
obtain the desired contaminant free surface.

An alternative to thermal vacuum cleaning, used  
by the prior art to effect cleaning of metal components,  
is solvent extraction. The solvent extraction cleaning  
25 process, in addition to requiring protracted treatment  
times, has the drawback that when cleaning porous  
materials, trace amount of the solvent used for  
cleaning, e.g., chlorinated hydrocarbons, may be  
adsorbed on the part being cleaned thereby, contri-  
30 buting to the contamination problem.



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SUMMARY OF THE INVENTION

In accordance with the present invention, the rapid removal of organic-based contaminants from articles, both porous and non-porous, without damage or contamination to the article is effected by contacting the article bearing the contaminant in a pressure vessel with a gas under super-critical conditions of temperature and pressure, whereby the contaminant on the surface and/or in the interstices of the article is absorbed by the gas and, thereafter, purging the gas from the pressure vessel to obtain the article having the contaminant removed therefrom. By surfaces is meant not only exterior surfaces but also interior surfaces which communicate therewith.

15

By the practice of the present invention, organic contaminants are removed from articles in one hour or less to achieve a substantially contaminant-free article as compared to thermal vacuum cleaning processes which require fifteen hours or more to achieve an equivalent level of cleanliness and several days of treatment by solvent extraction.

20

As will hereinafter be further demonstrated, by following the practice of the present invention, twice as much volatile contaminant was removed to temperatures near ambient from difficult-to-clean silicone rubber parts in one-fiftieth the time when compared to cleaning equivalent rubber parts using thermal vacuum cleaning.

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DETAILED DESCRIPTION OF THE INVENTION

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It is known that when the temperature of a gas is above a certain temperature, known as the critical temperature, it is not possible to liquefy the gas by application of pressure alone. It is necessary to reduce the temperature below the critical temperature in order to be able to liquefy the gas. At the critical temperature, as the gas is subjected to increasingly

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1 higher pressure, e.g., on the order of several thousand  
pounds per square inch (psi) (one psi equals 51.71493 mm  
of mercury), the density of the gas approaches that of  
a liquid and the gas acts as a solvent for a variety of  
5 different types of organic and organo-metallic materials,  
including aliphatic and aromatic hydrocarbon organo-  
metallics such as metal alkyls and alcoholates, silicones  
and boroalkyls and organic esters or inorganic acids  
such as sulfuric and phosphoric acid. The critical  
10 temperatures and pressures for a variety of gases at  
which they exist in the super-critical condition may be  
found in U.S. Patent No. 4,124,528, the teachings of  
which are hereby incorporated by reference.

15 In the practice of the present invention, the  
article of manufacture to be cleaned is placed in a  
suitable vessel such as a pressure chamber or autoclave  
and the gas which is to effect the cleaning of the  
article surface is admitted to the vessel in a super-  
critical condition.

20 Cleaning of the article is accomplished in the  
pressurized vessel under conditions which maintain the  
super-critical condition of the gas used for cleaning.  
Normally, the cleaning is conducted at a temperature  
range of about 35°C to about 100°C at about 1200 psi  
25 (62,058 mmHg) to about 10,000 psi (517,149 mmHg) pressure  
and preferably about 40°C to about 50°C and about 3,000  
psi (155,145 mmHg) to about 8,000 psi (413,719 mmHg)  
pressure. Inert gases having a critical temperature  
below about 200°C are considered most advantageous in  
30 the practice of the present invention. Examples of  
such gases are alkanes and especially lower alkanes  
such as ethane, propane and butane, alkenes and especially  
lower alkenes such as ethylene, propylene and butylene,  
dialkyl ethers such as dimethyl ether, SO<sub>2</sub>, CO<sub>2</sub>,

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1 halogenated alkanes such as  $\text{CHF}_3$ ,  $\text{CClF}_3$ ,  $\text{CFCI}_3$ ,  $\text{CF}_2=\text{CH}_2$ ,  
2  $\text{CF}_3$ ,  $\text{CF}_3\text{-CF}_2\text{-CF}_3$ ,  $\text{CF}_4$ ,  $\text{CH}_3\text{-CF}_3$ ,  $\text{CHCl}_2\text{F}$ ,  $\text{CCl}_2\text{F}_2$ ,  $\text{N}_2\text{O}$ ,  
3 noble gases such as argon,  $\text{NH}_3$  and  $\text{N}_2$ . Gases such as  
4  $\text{CO}_2$  are preferred in the practice of the present  
5 invention as the super-critical temperature of such  
6 gases is near ambient temperature; the gases are  
7 inexpensive, non-toxic, and relatively inert to most  
8 solid substrates.  $\text{CO}_2$  is especially preferred as this  
9 gas in the super-critical state has a very low viscosity,  
10 namely 0.05 centipoise, which is one-twentieth that of  
11 water. As a result, the gas in the super-critical  
12 state can penetrate very readily into the contaminant  
13 to effect its rapid removal from the article being  
14 cleaned.

15 To promote the rapid cleaning of the article  
16 with the super-critical gas, it is further advantageous  
17 to the practice of the present invention that the  
18 article to be cleaned be preheated prior to its place-  
19 ment in the pressure vessel to a temperature above  
20 ambient, e.g., about  $30^\circ\text{C}$  to about  $100^\circ\text{C}$ , and preferably  
21 about  $40^\circ$  to  $50^\circ\text{C}$ .

22 The absorptive capacity of the gases in the super-  
23 critical condition with respect to most contaminants,  
24 and particularly contaminants of basically organic  
25 origin, is raised with increased pressure. Thus, when  
26 practicing the cleaning procedure in accordance with  
27 the practice of the present invention, a pressure which  
28 is substantially higher than the critical pressure of  
29 the gas and a temperature only slightly above the  
30 critical temperature is selected for maintaining the  
31 gas in the super-critical condition.



1           It is still further advantageous to the practice  
of the present invention that the temperature and  
pressure conditions under which the gas is caused to  
contact the article to be cleaned be sufficiently  
5   above the critical temperature and pressure in order  
to have a single physical phase, i.e., the gaseous  
phase, of the gas present in the pressurized vessel  
during the cleaning operation. Thus, for CO<sub>2</sub> which  
has a critical temperature of 32°C and a critical  
10   pressure of 1073 psi (55,490 mmHg), when such gas is  
used as the cleaning medium, the gas is maintained at a  
temperature of about 35°C to about 100°C and a pressure  
of 2,000 psi (103,430 mmHg) to 10,000 psi (517,149 mmHg)  
in the pressure vessel.

15           In effecting cleaning of the surfaces of articles  
in accordance with the practice of the present invention,  
the article, when placed in the pressure vessel for  
cleaning, is contacted with the gas under super-critical  
conditions for a period of time ranging from about  
20   0.25 hour to about four hours and preferably about 0.5  
hour to about one hour to effect complete removal of  
contaminants.

          After sufficient time has elapsed in the pressure  
vessel for the contaminants to be absorbed by the gas  
25   and removed from the articles, the pressure in the  
vessel is released and the gas containing the absorbed  
contaminants are vented or purged from the vessel  
into the atmosphere. When ambient pressure is attained,  
the cleaned article is then removed from the vessel.

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1           If it is intended that the gas be recycled for  
reuse in removing contaminants, the gas in the super-  
critical condition is vented or purged from the  
pressurized vessel into a suitable collection vessel  
5       where the pressure is reduced or the temperature lowered  
at constant pressure, which conditions render the gas a  
non-solvent for the contaminant which then precipitates  
from the gas. The gas, freed of contaminants, can then  
be recompressed and recycled for use in the cleaning  
10      of contaminated articles.

          The following are examples showing the cleaning  
of various articles of manufacture using gases in the  
super-critical condition according to the method of  
the present invention.

15

Example I

          A high pressure autoclave (10,000 psi or  
517,149 mmHg maximum working pressure) of 300 milliliter  
(ml) capacity was equipped with a gas inlet, a gas  
20      outlet, pressure gauge, a thermocouple well, and heating  
means. Connected to the gas inlet was a CO<sub>2</sub> supply  
bottle which delivered the CO<sub>2</sub> at 800 psi (41,372 mmHg)  
gauge. A gas booster pump operating on the 100 psi  
(5171 mmHg) shop air and having the capability to raise  
25      the bottle pressure to a maximum to 10,000 psi  
(517,149 mmHg) was connected to the CO<sub>2</sub> bottle. The  
autoclave was purged with CO<sub>2</sub> and heated to 100°F  
(37.8°C). An O-ring formed of silicone rubber, weighing  
0.460 grams, was placed in the autoclave. CO<sub>2</sub> gas was  
30      fed to the booster pump and the autoclave was pressurized  
to 8,000 psi (413,719 mmHg).

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1           After contact of the O-ring with the CO<sub>2</sub> at  
100°F (37.8°C) and 8,000 psi (413,719 mmHg) pressure  
for one hour, the pressure in the autoclave was released  
and the CO<sub>2</sub> containing the absorbed contaminants was  
5           vented to the atmosphere. When ambient pressure was  
attained in the autoclave, the cleaned O-ring was  
removed from the autoclave and weighed to determine the  
extractable weight loss effected by the cleaning  
operation. The weight loss was determined to be 0.011  
10           grams which represented the removal of 2.4 weight  
percent (wt. %) contaminants.

          By way of contrast, an identical silicone rubber  
O-ring weighing 0.494 grams, was cleaned using an all  
glass thermal vacuum cleaning unit wherein the O-ring  
15           was heated for 120 hours and 180°C under a vacuum of  
approximately 10<sup>-5</sup> torr (mmHg). The extractable weight  
loss was determined to be 0.0014 grams, representing  
the removal of 0.4 wt. % contaminants. Subsequent  
contact of the thermal vacuum cleaned O-ring with  
20           isopropanol solvent at ambient laboratory temperature  
removed another 0.004 grams of contaminant or an  
additional 0.8 wt. % representing a total contaminant  
removal of only 1.1 wt. %.

          To determine the effect of the super-critical CO<sub>2</sub>  
25           treatment on the physical properties of the silicone  
rubber material, O-rings which had been treated in  
accordance with the procedure of Example I were subjected  
to tensile and hardness tests used for the evaluation  
of rubber mechanical and physical properties.

30           The results of these tests are recorded in  
Table I below.



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TABLE ITensile and Hardness Properties

5

Silicone Rubber Treatment	Tensile Strength* (psi) (mmHg)	Elongation* (%)	Hardness* (Shore A)
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Super-critical CO <sub>2</sub>	854 44,165	132	75
None	907 46,905	158	72

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\*Average of six tests

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The results recorded in Table I show that the treatment of silicone rubber with CO<sub>2</sub> under super-critical conditions produces only a minor change in the mechanical and physical properties of the rubber.

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Example II

The procedure of Example I was repeated to clean a polyimide polymer containing contamination in the form of volatile solvents by exposure to CO<sub>2</sub> for one hour under super-critical conditions of 8,000 psi (413,719 mmHg) pressure and a temperature of 45°C.

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The amount of volatile contaminants remaining in the polymer after cleaning was determined by the American Society for Testing and Materials (ASTM) TEST E-595, described in the 1981 Annual Handbook of ASTM Standards,

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1 under conditions of 125°C temperature and a vacuum of  
10<sup>-5</sup> torr (mmHg) or thermogravimetry mass spectrometry  
(TGA-MS) under conditions of one atmosphere and  
temperatures of 210°C or 820°C.

5 The results of these tests are recorded in Table II  
below.

For purposes of contrast, identical samples of  
the polyimide polymer were cleaned using thermal vacuum  
cleaning conditions wherein the polymer sample was  
10 heated for four hours at 80°C under a vacuum of approxi-  
mately 10<sup>-5</sup> torr (mmHg).

The results of these comparative tests are also  
recorded in Table II below.

15

TABLE II

Volatiles Remaining in Polyimide Polymer

20

Sample No.	Super-critical CO <sub>2</sub> Cleaning	Thermal Vacuum Cleaning	Analysis Conditions
1	0.56%	8.31%	ASTM E-595
2	0.76%	6.50%	ASTM E-595
3	1.00%	6.00%	TGA-MS, 210°C
4	37.00%	47.00%	TGA-MS, 820°C

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The reduction in volatiles is important in polymer  
processing as it suppresses the formation of voids and  
areas of weakness in the finished product.

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1           By reference to Table II, it is immediately apparent  
that by the practice of the present invention the removal  
of volatiles from polymeric products can be achieved to  
a much greater degree in a shorter period of time as  
5       compared to the practice of the prior art as represented  
by thermal vacuum cleaning.

Example III

10           The procedure of Example I was repeated with the  
exception that thin-sectioned parts of less than 0.25  
inch thickness of a diverse selection of organic and  
inorganic materials were cleaned by exposure to CO<sub>2</sub>  
under super-critical conditions with only minor changes  
15       in the mechanical and physical properties of the  
materials being observed thereafter.

The materials exposed to the super-critical CO<sub>2</sub>  
conditions were as follows:

- 20           A. Laser casting alloy.
- B. Fluorosilicone sheet.
- C. Glass reinforced epoxy resin  
              multilayer sheet.
- D. Fiberized carbon.
- E. Absorptive fabric containing activated  
25           carbon.
- F. Phenolic laminate cloth.
- G. Polyimide resin sheet.
- H. Quartz crystal assembly.
- I. Cryogenic cooler part.

30           The laser casting alloy was subjected to a vacuum-  
pressure cycle in silicone oil (Dow-Corning DC-200) to  
saturate the metal with the silicone oil. The oil-  
saturated metal part was then cleaned according to  
35       Military Interim Specification (MIS) 23542D, a cleaning  
specification for these parts. According to MIS-23542D,



1 the material to be cleaned is subjected to an exhaustive  
extraction in a Soxhlet apparatus using toluene as the  
solvent followed by evaporation of the solvent and an  
infrared (IR) spectra examination of the residue. In  
5 accordance with MIS-23542D, the IR examination  
must indicate the absence of silicone or other  
residues to establish removal of all traces of silicone  
oil contaminant. To achieve this result required four  
days of treatment with the Soxhlet extraction apparatus,  
10 whereas by using the procedure of Example I, removal  
of all traces of silicon oil contaminants from a similar  
laser casting alloy similarly saturated with silicone  
oil was achieved in two hours.

While specific components of the present system  
15 are defined above, many other variables may be introduced  
which may affect, enhance or otherwise improve the  
present invention. These are intended to be covered  
herein. Further, while variations are given in the  
present application, many modifications and ramifications  
20 will occur to those skilled in the art upon reading the  
present disclosure. These, too, are intended to be  
included herein.

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CLAIMSWhat is Claimed is:

- 1           1. A method of removing from a solid article  
contaminants adhering to the surfaces of the article,  
which comprises:  
          placing the article having contaminants  
5   affixed to the surfaces thereof in a pressure vessel;  
          introducing a gas into the vessel and  
maintaining the gas at a temperature and pressure at  
which the gas is placed in a super-critical condition;  
          maintaining the article in contact with the  
10   super-critical gas for a time sufficient to effect  
absorption of the contaminants by the gas; and  
          removing the gas containing the contaminants  
from the vessel, whereby the contaminants are separated  
and removed from the article.
- 1           2. The method of Claim 1 wherein the gas is  
carbon dioxide.
- 1           3. The method of Claim 2 wherein the carbon  
dioxide is maintained in the vessel at a temperature  
in the range of about 35°C to about 100°C and a  
pressure of about 1,500 psi (77,572 mmHg) to about  
5   10,000 psi (517,149 mmHg).
- 1           4. The method of Claim 1 wherein the contaminants  
are composed of organic-based materials.
- 1           5. The method of Claim 1 wherein the article is  
maintain d in contact with the gas in the super-critical  
state for a period of time ranging from about 0.25 to  
about four hours.



- 1           6. The method of Claim 1 wherein the article  
comprises a rubber.
- 1           7. The method of Claim 1 wherein the article  
comprises a metal.
- 1           8. The method of Claim 1 wherein the article  
comprises a synthetic organic polymer.
- 1           9. The method of Claim 1 wherein the article  
comprises a polyimide polymer.
- 1           10. The method of Claim 1 wherein the article  
comprises carbon.
- 1           11. The method of Claim 1 wherein the article  
comprises a quartz crystal.
- 1           12. The method of Claim 1 wherein the article  
comprises an assembly of parts.
- 1           13. The method of Claim 1 wherein the contaminant  
is removed from the external surface of the article by  
the gas.
- 
- 1           14. The method of Claim 1 wherein the contaminant  
is removed from the interstices of the article.





## AMENDED CLAIMS

(received by the International Bureau on 25 April 1984 (25.04.84))

1           1. A method of removing from a solid structural  
component or element contaminants produced during the  
manufacture and processing of the component or element  
and adhering to the surfaces of the component or element,  
5       which comprises:

          placing the component or element having  
contaminants affixed to the surfaces thereof in a  
pressure vessel;

          introducing a gas into the vessel and  
10       maintaining the gas at a temperature and pressure at  
which the gas is placed in a super-critical condition;  
          maintaining the component or element in  
contact with the super-critical gas for a time sufficient  
to effect absorption of the contaminants by the gas;  
15       and

          removing the gas containing the contaminants  
from the vessel, whereby the contaminants are separated  
and removed from the component or element.

1           2. The method of Claim 1 wherein the gas is  
carbon dioxide.

1           3. The method of Claim 2 wherein the carbon  
dioxide is maintained in the vessel at a temperature  
in the range of about 35°C to about 100°C and a  
pressure of about 1,500 psi (77,572 mmHg) to about  
5       10,000 psi (517,149 mmHg).

1           4. The method of Claim 1 wherein the contaminants  
are composed of organic-based materials.



1           5. The method of Claim 1 wherein the component  
or element is maintained in contact with the gas in the  
super-critical state for a period of time ranging from  
about 0.25 to about four hours.

1           6. The method of Claim 1 wherein the article  
comprises a rubber.

1           7. The method of Claim 1 wherein the article  
comprises a metal.

1           8. The method of Claim 1 wherein the article  
comprises a synthetic organic polymer.

1           9. The method of Claim 1 wherein the article  
comprises a polyimide polymer.

1           10. The method of Claim 1 wherein the article  
comprises carbon.

1           11. The method of Claim 1 wherein the article  
comprises a quartz crystal.

1           12. The method of Claim 1 wherein the article  
comprises an assembly of parts.

1           13. The method of Claim 1 wherein the contaminant  
is removed from the external surface of the component  
or element by the gas.

1           14. The method of Claim 1 wherein the contaminant  
is removed from the interstices of the component or  
element.



**STATEMENT UNDER ARTICLE 19**

The above-listed amendments are being submitted in accordance with the Patent Cooperation Treaty Article 19 and have been made for the following purposes.

Claim 1 has been amended to more particularly point out that the claimed process is used for the treatment of solid structural components or elements, as opposed to a particulate material, such as an adsorbent. In addition, Claim 1 has been amended to more particularly point out that the contaminants removed by the process of the present invention are contaminants produced during the manufacture and processing of the structural component or element, as opposed to an adsorbate which is intentionally adhered to an adsorbent as part of a purification process. These changes are deemed to clearly distinguish Applicant's claimed process over the references cited in the international search report.

Claims 5, 13, and 14 were each amended to provide language which is consistent with the language of Claim 1 from which these claims depend.



# INTERNATIONAL SEARCH REPORT

International Application No PCT/US 83/01713

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (if several classification symbols apply, indicate all) <sup>5</sup>		
According to International Patent Classification (IPC) or to both National Classification and IPC		
IPC <sup>3</sup> : B 08 B 7/00		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched <sup>4</sup>		
Classification System	Classification Symbols	
IPC <sup>3</sup>	B 08 B; B 01 D	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>5</sup>		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT</b> <sup>14</sup>		
Category <sup>6</sup>	Citation of Document, <sup>16</sup> with indication, where appropriate, of the relevant passages <sup>17</sup>	Relevant to Claim No. <sup>18</sup>
X	US, A, 4124528 (MODELL) 7 November 1978 see the entire document cited in the application --	1-14
X	US, A, 3969196 (ZOSEL) 13 July 1976 see the entire document -----	1-14
<p><sup>15</sup> Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&amp;" document member of the same patent family</p>		
<b>IV. CERTIFICATION</b>		
Date of the Actual Completion of the International Search <sup>1</sup>	Date of Mailing of this International Search Report <sup>2</sup>	
13th February 1984	02 MARS 1984	
International Searching Authority <sup>1</sup>	Signature of Authorized Officer <sup>19</sup>	
EUROPEAN PATENT OFFICE	G.L.M. Kruidenberg	

ANNEX TO THE INTERNATIONAL SEARCH REPORT ON

INTERNATIONAL APPLICATION NO.

PCT/US 83/01713 (SA 6067)

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 24/02/84

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